Mechanism and dynamics of biexciton formation from a long-lived dark exciton in a CdTe quantum dot

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Numerous optical studies of various quantum dot (QD) systems established a toolbox of techniques effective in characterization of single-dot photoluminescence (PL) spectra. In particular, measurement of the PL spectrum as a function of the excitation intensity is often employed to distinguish between spectroscopic lines related to different excitonic complexes [1]. The results are usually discussed in terms of a power-law behavior of the PL intensity. The simplistic stochastic model states for example that the formation of the biexciton requires a coincidence of two exciton formation events [2], and thus the biexciton PL intensity should increase quadratically with the excitation power. In real experiments, however, the biexciton PL intensity does not follow this prediction and exhibits less steep dependence [3].

Here we analyze the dependence of a single CdTe QD PL on pulsed excitation intensity and demonstrate contributions of two mechanisms of the biexciton formation: either from an empty dot by capture of two electron-hole pairs within a single excitation pulse or from a resident dark exciton created earlier [4]. We show that in the wide range of intensities the latter mechanism is dominant, which provides a natural explanation for subquadratic biexciton PL intensity power dependence. It is also a general example of the importance of the dark exciton state, which is often neglected [3,5], but should be taken into account in the rate-equation models to correctly describe the QD physics under various excitation regimes, including the CW excitation.

The discussed mechanism allows us to create the single biexciton with the use of two different laser pulses, polarization of which can be controlled independently. We exploit this possibility in the time-resolved experiments to study the impact of a carrier spin-blockade effect on the biexciton (and other excitonic complexes) formation dynamics. Our results show that the formation of the biexciton is slowed down when the two consecutive pulses used to create this complex have opposite circular polarization. Such effect is caused by relatively long relaxation time of a spin blockaded electron pair, while the spin-blockaded holes relax rapidly to their ground state.

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